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Title: Utilization of strawberry and raspberry waste for the extraction of bioactive compounds by deep eutectic solvents.

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ABSTRACT

Deep eutectic solvents were used for the recovery of bioactive compounds from strawberry and raspberry by-products. The best performing eutectic solvent (DES-7) was compared to one organic solvent. Total and individual phenolics, sugars, uronic acid, anthocyanins and the antioxidant activity *in vitro* were measured. The highest yield and purity of phenolic compounds in the extracts were obtained from strawberry extrudate with acetone (11.5 %) and from raspberry extrudate (36.7 %) with DES-7. The greatest yields of total sugars and acid sugars were achieved from raspberry extrudate using acetone and from strawberry extrudate using DES-7, 82.23 and 1.88 mg/g of extract, respectively. DES-7 was the solvent that extracted the most anthocyanins (1.28 mg/100 mg of extract). In addition, the extracts obtained with acetone presented the best values of antioxidant activity *in vitro* (DPPH value of 5.33 mmol TE/kg fresh weight and ORAC value of 37.89 µmol TE/g fresh weight) the extracts obtained with DES-7 also presented an interesting antioxidant activity, and a promising solvent for the extraction of bioactive compounds.

Keywords: Deep eutectic solvent, phenolics, anthocyanins, antioxidant activity, berry extrudate.

1. INTRODUCTION

The production of strawberries and raspberries has increased in recent years due to of a greater demand for these fruits because they are a rich source of essential nutrients such as fiber, potassium, folic acid and vitamin C. In the case of minor compounds, strawberries contain high levels of flavonoids, with anthocyanins being the most predominant type (Misran, Padmanabhan, Sullivan, Khanizadeh, & Paliyath, 2015). In addition to important concentrations of vitamins A and B, raspberries contain ellagitannins and anthocyanins, and are relatively high in iron (Bobinaitė, Viškelis, & Venskutonis, 2016). Through various studies it has been determined that the bioactive compounds present in these fruits, such as ellagic acid, have antioxidant (Sharma, Sharma, Singh Tuli, & Sharma, 2018), anti-inflammatory, and anticancer properties, (Giampieri et al., 2012).

The most important by-product generated from the berry industry is the so-called extrudate that still contains a large number of bioactive compounds of great interest, such as phenolics like ellagic acid which has anticarcinogenic properties (Giampieri et al., 2012). Moreover, the strawberry's achenes would remain in the extrudate, which, in spite of being a minor part of the fruit's composition, contribute to more than 41% of the total antioxidant content and represent 81% of this antioxidant capacity (Ariza et al., 2016). Thus, the poor management of this by-product would mean a substantial loss of bioactive compounds.

Several studies have been carried out in which strawberry extrudate was used to favor the biomethanation of sewage sludge, as this berry residue biodegrades with greater ease and enables the dilution of the inhibitors and contaminants that are found in the sludge, such as nitrogen and heavy metals (Serrano Moral, 2015). Despite this potential use, there are many interesting compounds which are beneficial to human health that should be recovered from the extrudate beforehand, such as organic acids, phenolics, anthocyanins or sugars (Ariza et al., 2016). This would fulfill a double function: on the one hand, bioactive compounds would be recovered for use in food formulations, and on the other hand, removing these same compounds would help subsequent treatment of the waste via bioprocesses to obtain energy, compost or for complete biodepuration.

To the best of our knowledge, only one study has explored the recovery of bioactive compounds from strawberry extrudate and none exist for raspberries. In the previously

published study, extractions at different temperatures were tested, and the use of a hydrothermal treatment at 150 ºC was found to be the most appropriate (Rodríguez-Gutiérrez et al., 2018). However, more literature exists on the analytic recovery from whole fruits, from which high amounts of phenolic compounds can be obtained using 70% acetone in water (Aaby, Ekeberg, & Skrede, 2007), or 80% methanol in water, for the specific extraction of anthocyanins and derivatives of hydroxycinnamic acid (Macheix, Fleuriet, & Billot, 1990). These extraction methods are fundamentally analytical in nature, so it is necessary to look for new extracting agents that allow a substantial amount of bioactive compounds to be recovered from strawberry and/or raspberry extrudates in a more sustainable way.

Deep eutectic solvents (DES) are "green" solvents which are analogous of ionic liquids and are systems formed from the mixture of Brønsted-Lowry acids and bases and may contain a variety of cationic and/or anionic species. They are generally obtained by the complexation of a quaternary ammonium salt with a metal salt or a hydrogen bond donor. DES have advantages over other organic solvents, such as high biodegradability and ease of handling due to the low toxicity they present (Ruesgas-Ramón, Figueroa-Espinoza, &Durand, 2017). Higher yields of oleacein and oleocanthal can be obtained from olive oil by using DES as extracting agent than when using methanol in water (García, Rodríguez-Juan, Rodríguez-Gutiérrez, Rios, & Fernández-Bolaños, 2016). In other studies DES have been used for antioxidant extraction from onions (Pal &Jadea, 2019) or rutin and quercetin from plants (Ma, Tang & Row, 2017) or even phenolics and furanocoumarins from leaves (Wang, Jiao & Gai, 2017).

However, to date, no studies exist on the use of these solvents for the extraction of bioactive compounds from strawberries or raspberries.

The main objective of this work was to compare the extraction of bioactive compounds from berry extrudates using DES with the commonly used organic solvents to make the utilization of these wastes more sustainable. Secondary objectives included determination of the compounds of interest extracted and their *in vitro* antioxidant activity.

2. MATERIALS AND METHODS

2.1. Materials

The fruit extrudates employed were supplied by the company HUDISA S.A., Lepe (Huelva, Spain). The strawberry extrudate was supplied from the 2016-2017 season; the raspberry extrudate was supplied from the 2017-2018 season.

The samples were stored at -20 ºC before treatment and analysis, and were thawed at 4 ºC the day before the treatments.

2.2. Extraction methods

In method 1 , the extracting agent used was acetone and the protocol described by Aaby, Ekeberg and Skerde (2007) was followed, in which 5 mL of the solvent were added to 5 grams of extrudate and the mixture was stirred at 4 ºC for 10 minutes.

In method 2 (DES), the extracting agents used were deep eutectic solvents. 5 mL of each eutectic solvent were added to 5 grams of extrudate, and stirred for 15 minutes. The mixture was centrifuged for 10 minutes at 1765 g. The supernatant was filtered using glass wool, and then centrifuged at 13710 g for 10 minutes.

2.3. Chemicals

Gallic acid (GA) standards, Folin-Ciocalteu's phenol reagent, anthrone and 2,2-diphenyl-1-picrylhydrazyl were purchased from Sigma-Aldrich (Madrid, Spain). Sodium bicarbonate was from PanreacQuimica S.A. (Barcelona, Spain). Acetone was obtained from PanReac AppliChem, the glacial acetic acid came from EMPARTA® ACS, and the methanol from Honeywell. Ultrapure water was obtained from a Milli-Q water system (Millipore, Milford, MA, USA).

2.4. DES preparation

To obtain the eutectic solvents, the starting components were mixed, as indicated in Table 1. In a round-bottomed flask, the corresponding mixtures of the DES were heated to 60° C with agitation in a rotary evaporator (without vacuum) until the formation of the solvent, which was a viscous, colorless and stable liquid. However, for those solvents containing sucrose in their composition (DES-2 and DES-5), all the compounds were previously dissolved with the necessary amount of water; and the excess water was eliminated in a rotary evaporator with vacuum until their formation.

2.5. Chromatographic determination and identification of phenolic compounds

The content of phenolic compounds was determined using the HPLC method. A Beckmn Coulter system with liquid chromatography equipment and a System Gold 168 detector, using a Merck Superspher C18 reverse phase column (250 mm x 4 mm), was used for the verification of the phenolic compounds. The mobile phases were water (A) (2.5% formic acid) and methanol (B) (2.5% formic acid), following the method of Gil, Holcroft, & Kader, 1997 as modified by Pérez, García-Rodríguez, Sanz, & Refoyo, 2017**.** The gradient was linear from 15 to 30% B in the first 15 min, and held for 5 min. After that, the gradient was from 30 to 80% B for 5 min, and finally an isocratic mixture for 2 min before returning to the initial conditions. The flow rate was 1 mL/min and the wavelength of the detector was performed at 280, 320, 350, and 510 nm.

2.6. Determination of the amount of dry extract

The amount of extract obtained from the extrudates was gravimetrically determined. Between 0.5 mL and 2 mL of extract were air-dried and introduced into a desiccator until a constant weight was reached, with the results expressed as milligrams of extract per gram of fresh sample. For samples extracted using eutectic solvent, the dry residue of each eutectic solvent was determined to correct the gravimetric measure.

2.7. Total soluble phenolic compounds

Total soluble phenolics were determined using the Folin-Ciocalteu method (Singleton & Rossi, 1965). Absorbance was measured at 655 nm and the results were expressed as micrograms of gallic acid equivalents per gram of fresh weight.

2.8. Total soluble anthocyanins

Total soluble anthocyanins were quantified following a previously described protocol (Serrano Mula, 2013), with some modifications. Briefly, the sample was mixed with a volume of 80% MeOH:H_2O in a 1:2 ratio, using ultraturrax homogenization equipment (IKA, Germany). The mixture was centrifuged, at 1765 g for 10 minutes and then at 13710 g for 10 minutes. Absorbance was read at 529 nm for a sample volume in a microplate (in duplicate) and the same volume of 80% MeOH:H_2O mixture was used as a blank.

The amount of anthocyanins (Ant.), expressed as mg cyanidin-3-glucoside equivalents per gram of fresh weight (mg c/g), was determined by the expression:

$$\text{Total Ant.}\left(\frac{\text{mg c}}{\text{g}}\right) = \frac{Abs\ 529\ nm x V(L) x\ 449.6\frac{g}{mol}(Reference\ anthocyanin\ molecular\ weight) x\ 1000\frac{mg}{g}}{Weight(g) x\ 26900\frac{L}{cmxmol} x\ 1\ cm}$$

2.9. Total soluble sugars

Total soluble sugars were determined using the anthrone method (Mokrash, 1954). Absorbance was measured at 630 nm and the results were expressed as milligrams of glucose equivalents per gram of fresh weight.

2.10. Total soluble acid sugars

Total soluble acid sugars were determined using the m-hydroxybiphenyl chromogen method (Blumenkrantz & Asboe-Hansen, 1974). Absorbance was measured at 540 nm and the results were expressed as milligrams of galacturonic acid equivalents per gram of fresh weight.

2.11. Determination of free radical scavenger capacity (DPPH)

The determination of the free radical scavenger capacity was carried out following the DPPH assay (Sánchez-Moreno, Larrauri, & Saura-Calixto, 1998). Absorbance was measured at 490 nm and the results were expressed as an EC_{50} (effective concentration) in millimol of Trolox equivalents per kilogram of fresh weight.

2.12. Determination of the reducing power

Determination of the reducing power was carried out using the ferric reducing antioxidant power (FRAP) assay (Benzie & Strain, 1996). Absorbance was measured at 490 nm and the results were expressed as millimol of Trolox equivalents per kilogram of fresh weight.

2.13. Oxygen radical absorbance capacity (ORAC)

Determination of the oxygen radical absorbance capacity was carried out using the ORAC assay, following a previously described protocol (Ou, Hampsch-Woodill, & Prior, 2001) with modifications. Fluorescence was measured using excitation and emission wavelengths of 485 nm and 538 nm, respectively, with a measurement interval of 5 minutes for a total reading time of 90 minutes.

192 The area under the curve was determined for all the samples, according to the following
193 expression: A = (0.5 + f5 / f0 + f10 / f0 ... + f90 / f0) x int

194 Where A was the area under the curve, f0 was the fluorescence value at time zero, fi
195 was the value of the fluorescence at the different times and int was the time interval at
196 which each measurement was made.

197 A calibration line was obtained for the area values of the different Trolox solutions. The
198 final result was expressed as millimol of Trolox equivalents per kilogram of fresh
199 weight.

200 **2.14. Statistical analysis**

201 The One Factor Analysis of Variance (ANOVA) was performed to compare a number
202 of groups in a quantitative variable (Bakieva, González Such, & Jornet, 2012). To
203 determine whether the means differed from one another, a so-called post hoc or a
204 posteriori multiple comparison, such as the Tukey method, was used.

205 Correlations among variables were determined by the Pearson correlation coefficient
206 (Puth, Neuhäuser, & Ruxton, 2014).

207

208

209 **3. RESULTS AND DISCUSSION**

210

211 **3.1. Assays with different DES**

212 The effectiveness of different eutectic solvents (DES-1 to DES-7) was tested by
213 determining the soluble phenolic compounds and soluble anthocyanins present in
214 strawberry and raspberry extrudates, as well as their antioxidant activity by the DPPH
215 method (Table 2). The phenolic compounds of each extract were also analyzed by
216 chromatographic determination (Table 3).

217 The results obtained in both extrudates for total soluble phenolics and total soluble
218 anthocyanins (Table 2) showed that the highest concentrations of these compounds were
219 obtained using solvent DES-7. In the case of total phenolics, extractions from

220 strawberry and raspberry extrudates yielded different results for different solvents, with
221 the exception of DES-7, which seems to indicate that the extractability and phenolic
222 composition of both extrudates must be different. Higher amounts of total phenolics
223 were extracted in the case of strawberry, reaching almost twice that of raspberry with
224 (the same?) DES. A higher content of total anthocyanins was also extracted from
225 strawberry, with particularly high levels achieved from both extrudates with the use of
226 solvent DES-7.

227 The results of the DPPH assay for the strawberry extrudate showed no significant
228 differences among the uses of DES-1, DES-6 and DES-7 ($p > 0.05$), but in the case of
229 the raspberry extrudate, the highest value for antioxidant activity was obtained with
230 DES-7 (1.68 mmol Trolox equivalents/kg fresh weight). Due to the greater extraction of
231 phenolics, including anthocyanins, the results of the DPPH assay showed higher values
232 for the extracts obtained from the strawberry extrudate (2.19-2.27 mmol Trolox
233 equivalents/kg fresh weight).

234 From the HPLC analysis, higher concentrations of quercetin-3-glucoside, ellagic acid
235 derivative and kaempferol (27.94, 21.95 and 34.26 µg/g fresh weight, respectively)
236 were obtained from the strawberry extrudate using the eutectic solvent DES-6;, while
237 higher concentrations of pelargonidin-3-glucoside and ellagic acid were obtained using
238 the solvent DES-7 (21.21 µg/g fresh weight) (Table 3). In general, a higher
239 concentration of phenolic compounds was obtained from extraction with DES-7, which
240 was in agreement with the results observed using the Folin-Ciocalteucolorimetric test.
241 Comparing the values obtained from each compound with previously published data on
242 whole fresh fruit (Aaby, Mazur, Nes, & Skrede, 2012; Tumbas Šaponjac et al., 2015), in
243 certain cases the amount obtained experimentally was greater than expected: 2479% in
244 the case of ellagic acid using solvent DES-7; and 3263% in the case of kaempferol with
245 solvent DES-6.

246 In the case of the raspberry extrudate, the concentration of the individual phenolic
247 compounds depended on the DES used. Comparing the values obtained in this study
248 with previously published data on whole fresh fruit (Burton-Freeman, Sandhu, &
249 Edirisinghe, 2016; Rommel & Wrolstad, 1993), in certain cases the amount obtained
250 experimentally was greater than expected bibliographically, especially in the case of
251 ellagic acid, which was 298% higher using DES-3, and in the case of kaempferol,

252 1530% higher with DES-6. Unlike for strawberry, the total phenolic concentrations
253 obtained from the raspberry extrudate using different DES were similar, ranging
254 between 26.5 and 36.8 %.

255

256 In the strawberry extrudate, the correlation coefficient obtained between the total
257 soluble phenolics and the DPPH stood out (0.75) (Table 4), which could be because a
258 large part of the antioxidant activity of the strawberry extrudate was due to its phenolic
259 concentration. Taking into account the chromatographically studied compounds, it is
260 worth highlighting the high correlation between pelargonidin-3-glucoside and total
261 soluble phenolics (0.77), which was due to the fact that pelargonidin-3-glucoside is the
262 major phenolic compound in strawberries. It also had a high correlation with
263 anthocyanins (0.76), as this compound is an anthocyanin derivative. Also noteworthy is
264 the high correlation between the total of chromatographically determined phenolic
265 compounds and DPPH (0.87), which showed the contribution of these compounds to
266 antioxidant activity, as well as the high correlation between phenolics and ellagic acid
267 (0.89), since the greatest concentration of this phenolic compound was obtained.

268 In the case of raspberry extrudate, there was a high correlation between the total
269 phenolics and the anthocyanins (0.81), and between anthocyanins and DPPH (0.90). It
270 is also worth mentioning that a high correlation between ellagic acid and ellagic acid
271 derivative was revealed from the chromatographic analysis (0.76), along with a high
272 correlation between the former and the total phenolic compounds determined
273 chromatographically, since the highest concentration of ellagic acid was obtained
274 (0.97).

275 **3.2. Comparison of extractions with DES-7 and analytical solvents for extraction**

276 As the solvent that provided the best extraction results was DES-7, solvent DES-7 was
277 employed for comparisons with other more conventional solvents used in berries, such
278 as acetone (Table 5), using the same extrudates as for extractions with deep eutectic
279 solvents.

280 *Phenolics and Anthocyanins*

281 It is important to note that the comparison of DES was carried out with fresh samples,
282 and the rest of the study was carried out using thawed samples. The higher extraction of
283 phenolics in Table 5 shows better extraction after the freezing process.

284 In the case of the strawberry extrudate (Table 5), acetone extraction led to a
285 significantly greater amount of total soluble phenolics (7157 μg/g).

286 In studies carried out with different strawberry varieties, the phenolic concentration was
287 determined to be between 2900 and 10200 μg of gallic acid/g whole fresh fruit (Lester,
288 Lewers, Medina, & Saftner, 2012). These values are close to those obtained for the
289 extrudate extracts in this work (322-7157 μg of gallic acid equivalents/g fresh weight),
290 and highlight the large amount of phenolic compounds remaining in the extrudate.

291 With respect to raspberry extrudate, the extract with the greatest amount of soluble
292 phenolic compounds was obtained when the eutectic solvent was used (49644μg/g), with
293 almost 160% higher than for extraction with acetone. This fact illustrates the advantage
294 of DES as an extracting agent of phenolic compounds from raspberry extrudate. It is
295 important to note that seven times more phenolics were recovered from raspberry
296 extrudate than from strawberry extrudate.

297 In previous studies, the concentration of total phenolics in raspberry fruit was
298 determined as approximately 15730 μg/g sample (Sette, Franceschinis, Schebor, &
299 Salvatori, 2017), so the fact that a higher concentration of these compounds was
300 obtained indicated that the solvents used to extract these compounds had high
301 extraction capacity (49644 μg of gallic acid equivalents/g fresh weight for DES-7).

302 The extraction of anthocyanins was higher from strawberry extrudate, with more than
303 double the amount extracted compared to raspberry extrudate with DES-7, the best
304 solvent used. As they are water-soluble compounds, the amount extracted from the
305 extrudates (2.83 and 1.28 mg of total anthocyanins/100g fresh sample, in strawberry and
306 raspberry extrudates, respectively) will always be below the concentration of
307 anthocyanins present in whole fresh fruits -- approximately 12 mg/100 g strawberries
308 (Panico et al., 2009) and 12.4-69.5 mg cyanidin-3-glucoside/100 g in raspberries
309 (Sariburun, Şahin, Demir, Türkben, & Uylaşer, 2010).

310 In general, a higher concentration of the individual phenolic compounds was obtained
311 using acetone. However, higher concentrations of ellagic acid than would be expected

for the whole fresh fruit (Pérez et al., 2017) were obtained using DES-7 (54.4 µg/g), with the economic and environmental advantages that this entails.

The results obtained chromatographically for raspberry extrudate showed that a higher concentration of phenolic compounds was obtained using acetone, as was the case for strawberry extrudate. For both extractions with acetone (67.2 µg/g) and DES-7 (54.4 µg/g), higher percentages of ellagic acid were obtained from the raspberry extrudate than those previously published (more than 100% in the acetone extract) (Burton-Freeman et al., 2016; Rommel & Wrolstad, 1993),

Sugars

The extraction methods with acetone did not guarantee the maximum recovery of sugars, since they were focused on the extraction of phenolic compounds.

The solvent DES-7 yielded the greatest amount of sugars from the strawberry extrudate (82.23 mg/g). The quantified sugars are the remains of soluble sugars present in the samples after the extrusion process plus the possible cell wall sugars that may be solubilized by the solvents used, hence the great difference in the results obtained using the different extraction methods. Therefore, the reason why the concentration was highest using DES-7 was because this solvent solubilized the most sugars from the cell wall material, something that was not observed for raspberry extrudate.

The sugar content in whole fresh strawberry samples was previously reported as 73.23 mg glucose/g fresh sample (Zeliou, Papasotiropoulos, Manoussopoulos, & Lamari, 2018), a value very similar to that obtained in this work using DES-7 (82.23 mg glucose equivalents/g fresh weight). For the raspberry extrudate, the acetone solvent extracted the highest content of sugars (39.95 mg/g). Previous studies determined the amount of sugars in raspberries as 4.4% (United States Department of Agriculture, 2018), expressed as percentage of glucose, a very similar concentration to that obtained with acetone in this work (3.99%).

In both cases, it should be noted that the sugars detected were free sugars, and that future studies on the use of these by-products should also take into account the wall sugars, such as hemicelluloses and cellulose.

Uronic acids

For the strawberry extrudate, a greater quantity of uronic acids was determined in the extract obtained using DES-7 (1.88 mg/g). This results indicated that the extract obtained by the eutectic solvent had a greater amount of pectins than the analysis carried out by other authors (Kintner & Buren, 1982). For the raspberry extrudate, the extract with the highest concentration of acid sugars was obtained using acetone (0.36 mg/g), in accordance with previous work. This result was also expressed for dry matter (Table 5), showing the selectivity and purity of each extraction method.

A greater percentage of total phenolics was obtained from the raspberry extrudate than the strawberry extrudate, particularly for DES-7 (36.53%). The extracts with the highest percentages of sugars were obtained using acetone in both extrudates (65.35-67.23%). Extraction with acetone solvent extracted mainly sugars and phenolics.

3.3. Determination of antioxidant activity *in vitro*

The extract obtained by acetone showed the highest antiradical DPPH activity for both extrudates (5.33 and 2.67 mmol TE/kg fresh weight for strawberry and raspberry extracts, respectively) (Table 5). It should be noted that the results for antioxidant activity in the strawberry extrudate were in agreement with those of soluble phenolic compounds.

In previous studies the antioxidant activity of strawberry extracts using the DPPH method was approximately 2.70 mmol of Trolox equivalents/kg fresh fruit weight (Voca et al., 2010). For raspberries, the antioxidant capacity was determined to be 2.6 mmol of Trolox equivalents/kg fresh fruit weight (Trivedi, Verma, & Tyagi, 2016). The values for both fruits were similar to those obtained in this work for some of the extracts (2.39-5.33 and 2.07-2.67 mmol of Trolox equivalents/kg fresh weight, in strawberry and raspberry extrudates, respectively). This is a striking result since a large amount of antioxidants were removed from the aqueous fraction during extrusion.

The extract obtained using acetone was the only one to have reducing power activity in the strawberry extrudate, while the other samples had no reducing power in the concentrations analyzed. In the raspberry extrudate, the sample with the highest reducing power activity was the extract obtained using acetone (335.60 millimol of Trolox equivalents per kilogram of fresh weight).

For the ORAC assay, the extract from strawberry extrudate with the highest concentration was obtained using acetone (37.89 mmol TE/kg fresh weight). In previous studies, where this test was performed on fresh strawberry samples at different stages of maturation (Wang & Lin, 2000), the concentration ranged from 9.7-21.3 mmolTrolox equivalents/kg fresh sample, lower than the values obtained in this work (1.89-37.89 mmol Trolox equivalents/kg fresh weight).

For the raspberry extrudate, the extract with the highest concentration was obtained using acetone (19.70 mmol TE/kg fresh weight). Previous studies (Wang & Lin, 2000) determined that at different maturation stages, the concentration for red raspberries was 10.9-18.2 mmol Trolox equivalents/kg fresh sample, similar to the values obtained in this work (1.36-19.70 mmol Trolox equivalents/kg fresh sample).

3.4. Correlation between components and activities

In the case of the strawberry extrudate (Table 6), high correlation coefficients between DPPH and the assays of total phenolic compounds (0.92) and reducing power (0.80) were obtained, with values very close to 1, indicating a close relationship between these variables. A high correlation was also found between soluble acid sugars and anthocyanins (0.96), and a medium correlation between soluble sugars and acid sugars, phenolics and anthocyanins. Among antioxidant methods, a high correlation was found for DPPH and ORAC.

For the raspberry extrudate (Table 6), a high correlation was observed between total phenolics and anthocyanins (0.95), with anthocyanins constituting approximately 30% of the total phenolic compounds, as previously described. The results obtained by the DPPH, reducing power and ORAC methods were related. Unlike the strawberry, there was an inverse correlation between total sugars and anthocyanins or total phenolics.

4. CONCLUSIONS

The use of eutectic solvents like DES-7 as extracting agents resulted in a high recovery of phenolics, anthocyanins and sugars from both extrudates. The extract obtained presented antioxidant activity *in vitro* with the DPPH, reducing power (FRAP) and

ORAC methods.

In view of the results obtained, acetone and the deep eutectic solvent DES-7 stand out as the best extracting solvents for the recovery of these compounds. On the one hand, this study confirmed the effectiveness of acetone for the extraction of bioactive compounds from strawberry and raspberry extrudates, as reported in the literature for whole fresh fruits. On the other hand, this study highlighted the effectiveness of the new eutectic solvents, in this case DES-7, for the extraction of bioactive compounds from these fruits. The use of eutectic solvents for extraction, as alternatives to organic solvents such as acetone, is of great interest, particularly in relation to the environment, since they are not as polluting or damaging to human health. In addition, these types of solvents can be used as vehicles for the solubilized bioactive compounds; whereas other organic solvents cannot. For this reason, in the future, eutectic solvents may be the best method for the extraction of bioactive compounds from strawberry and raspberry extrudates.

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573 Table 1. Composition of the deep eutectic solvents used.

Abbreviation	Components			Molar ratio	% H_2O
DES-1	Choline chloride	Glycerol	-	1:2	-
DES-2	Choline chloride	Sucrose	-	1:2	25
DES-3	Choline chloride	1,4-Butanediol	-	1:5	-
DES-4	Choline chloride	1,2-Propanediol	-	1:1	7.5
DES-5	Betaine	Sucrose	-	2:1	13
DES-6	Betaine	Levulinic acid	-	1:2	-
DES-7	Choline chloride	Glycolic acid	Oxalic acid	1:1.7:0.3	-

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575

576 Table 2. Soluble phenolics, anthocyanins and antioxidant activity by DPPH method of
577 the extracts obtained using the different deep eutectic solvents from strawberry and
578 raspberry extrudate. Different letters indicate that there are significant differences
579 among the results of the extracts within the same extrudate ($p < 0.05$).

Deep EutecticSolvents	Total soluble phenolics		Total soluble anthocyanins		A
	µg gallic acid equivalents/g fresh weight		mg cyanidin-3-glucoside equivalents/100 g fresh weight		mmolTr
	Strawberryextrudate	Raspberryextrudate	Strawberryextrudate	Raspberryextrudate	Strawber
DES-1	4369.00 ± 6.33 d	2693.06 ± 120.28 B	0.35 ± 0.02 b	0.13 ± 0.05 B	2.19
DES-2	4085.46 ±166.71 d	1991.65 ±31.75 C	0.30 ± 0.05 b	0.06 ± 0.00 B	1.22
DES-3	3479.21 ±39.69 e	2131.98 ± 39.69 C	0.42 ± 0.03 b	0.07 ± 0.00 B	1.57
DES-4	5001.69 ±39.98 b	1837.91 ±78.63 C	0.21 ± 0.00 c	0.09 ± 0.01 B	1.78
DES-5	3293.96 ± 47.63 e	2581.06 ±134.96 B	0.34 ± 0.02 b	0.04 ± 0.00 B	1.39 ±
DES-6	5056.58 ± 63.51 b	2126.37 ±15.88 C	0.36 ± 0.00 b	0.09 ± 0.00 B	2.21
DES-7	5791.94 ±214.34 a	3355.71 ± 7.94 A	1.38 ± 0.11 a	0.38 ± 0.03 A	2.27

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582

583 Table 3. Concentration of phenolic compounds in the extracts obtained using the different deep eutectic solvents, using a HPLC system.
584 Different letters indicate that there are significant differences among the results of the extracts within the same compound ($p < 0.05$).

Deep Eutectic Solvents	µg phenol compound/g fresh weight											
	Pelargonidin-3-glucoside		Quercetin-3-glucoside		Ellagic acid derivative		Ellagicacid		Kaempferol		Sum of phenoliccompounds	
	Strawberryextrudate	Raspberryextrudate	Strawberryextrudate	Raspberryextrudate	Strawberryextrudate	Raspberryextrudate	Strawberryextrudate	Raspberryextrudate	Strawberryextrudate	Raspberryextrudate	Strawberryextrudate	Raspberryextrudate
DES-1	7.13 ± 0.05 c	7.49 ± 0.40 c	22.23 ± 4.14 c	5.09 ± 0.22 a	16.44 ± 2.55 d	2.68 ± 0.11 a	98.24 ± 7.12d	12.40 ± 2.55 a	22.15 ± 2.45 e	0.46 ± 0.03 a	166.2 ± 12.50 f	28.12 ± 2.13 a
DES-2	1.24 ± 0.07 b	7.31 ± 0.71 c	14.67 ± 2.16 b	4.48 ± 0.56 a	9.35 ± 1.06 c	2.63 ± 0.28 a	27.52 ± 5.92 b	11.08 ± 0.79 a	13.3 ± 0.93 d	1.01 ± 0,09 b	66.09 ± 5.45 c	26.52 ± 1.44 a
DES-3	1.19 ± 0.06 b	1.69 ± 0.09 b	19.56 ± 1.77	4.11 ± 0.40 a	16.03 ± 2.00	2.87 ± 0.15 a	79.72 ± 6.42 d	27.11 ± 1.80 b	20.67 ± 3.17 e	1.05 ± 0.05 b	137.17 ± 9.90 e	36.79 ± 1.99 b
DES-4	12.06 ± 1.44 d	7.69 ± 0.12 c	21.51 ± 3.29 c	4.06 ± 0.35 a	15.63 ± 1.44 cd	2.56 ± 0.17 a	42.09 ± 4.44 c	16.08 ± 2.21 a	21.53 ± 1.91 e	0.81 ± 0.01 a	112.73 ± 7.65 d	31.2 ± 2.04 ab
DES-5	0.50 ± 0.02 a	5.96 ± 0.31 c	10.79 ± 2.40 b	4.12 ± 0.27 a	5.22 ± 1.20 b	2.82 ± 0.09 a	36.24 ± 2.64 c	13.72 ± 1.00 a	3.89 ± 0.05 c	0.77 ± 0.01 a	56.64 ± 4.24 c	27.39 ± 1.00 a
DES-6	0.25 ± 0.02 a	0.35 ± 0.01 a	27.94 ± 4.05 c	4.48 ± 0.40 a	21.95 ± 3.18 d	3.04 ± 0.30 a	47.69 ± 2.75 c	26.98 ± 2.14	34.26 ± 3.70 f	1.53 ± 0.03 b	132.09 ± 10.06 e	36.39 ± 2.65 b
DES-7	21.21 ± 3.09 e	8.87 ± 0.98 cd	8.88 ± 1.28 b	4.42 ± 0.38 a	5.00 ± 0.10 b	2.76 ± 0.24 a	128.89 ± 9.10 e	13.71 ± 0.97 a	15.43 ± 1.15 d	0.68 ± 0.01 a	179.41 ± 11.78 f	30.45 ± 2.09 a

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586

Table 4. Pearson correlation coefficients among the different variables studied for the extracts obtained using deep eutectic solvents. The values marked in bold are equal to or greater than 0.40 in absolute value.

	Strawberry extrudate								
	Total soluble Phenolics	Anthocyanins	DPPH	Pelargonidin- 3-glucoside	Quercetin-3-glucoside	Ellagic acid derivative	Ellagicacid	Kaempferol	Sum of phenoliccompounds
Total soluble Phenolics	**1.00**	**0.59**	**0.75**	**0.77**	0.09	0.09	**0.45**	**0.46**	**0.62**
Anthocyanins	**0.59**	**1.00**	**0.49**	**0.76**	**-0.57**	**-0.51**	**0.79**	-0.14	**0.58**
DPPH	**0.75**	**0.49**	**1.00**	**0.55**	0.34	0.35	**0.69**	**0.62**	**0.87**
Pelargonidin-3-glucoside/ Cyanidin-3-glucoside	**0.77**	**0.76**	**0.55**	**1.00**	-0.38	-0.35	**0.69**	-0.05	**0.61**
Quercetin-3-glucoside	0.09	**-0.57**	0.34	-0.38	**1.00**	**0.99**	-0.22	**0.88**	0.22
Ellagicacidderivative	0.09	**-0.51**	0.35	-0.35	**0.99**	**1.00**	-0.14	**0.91**	0.29
Ellagicacid	**0.45**	**0.79**	**0.69**	**0.69**	-0.22	-0.14	**1.00**	0.11	**0.89**
Kaempferol	**0.46**	-0.14	**0.62**	-0.05	**0.88**	**0.91**	0.11	**1.00**	**0.53**
Total of phenoliccompounds	**0.62**	**0.58**	**0.87**	**0.61**	0.22	0.29	**0.89**	**0.53**	**1.00**
	Raspberryextrudate								
Total soluble Phenolics	**1.00**	**0.81**	**0.61**	**0.42**	0.34	0.09	-0.35	**-0.52**	-0.25
Anthocyanins	**0.81**	**1.00**	**0.90**	**0.45**	0.19	-0.06	-0.23	-0.34	-0.03
DPPH	**0.61**	**0.90**	**1.00**	**0.40**	0.23	-0.10	-0.14	-0.25	0.08
Cyanidin-3-glucoside	**0.42**	**0.45**	**0.40**	**1.00**	0.20	**-0.84**	**-0.93**	**-0.80**	**-0.81**
Quercetin-3-glucoside	0.34	0.19	0.23	0.20	**1.00**	-0.07	-0.31	-0.29	-0.30
Ellagicacidderivative	0.09	-0.06	-0.10	**-0.84**	-0.07	**1.00**	**0.76**	**0.67**	**0.67**
Ellagicacid	-0.35	-0.23	-0.14	**-0.93**	-0.31	**0.76**	**1.00**	**0.72**	**0.97**
Kaempferol	**-0.52**	-0.34	-0.25	**-0.80**	-0.29	**0.67**	**0.72**	**1.00**	**0.63**
Sum of phenoliccompounds	-0.25	-0.03	0.08	**-0.81**	-0.30	**0.67**	**0.97**	**0.63**	**1.00**

593 Table 5. Total soluble phenolics and individuals, anthocyanins, total sugars, acid sugars
594 and the *in vitro* antioxidant activity assays of the extracts obtained from strawberry
595 extrudate (SE) and raspberry extrudate (RE). Bioactive compounds refer to dry extract
596 for both extrudates (%). Different letters indicate significant differences among the
597 results of the extracts within the same extrudate ($p < 0.05$).

	Sample	Amount	Method 1 (acetone)	Method 2 (DES-7)
Total soluble phenolics	SE	µg/g	7157 ±160 a	5761±12 b
		%	11.50	1.99
	RS	µg/g	19405±377 B	49644±760 A
		%	31.74	**36.53**
Total soluble anthocyanins	SE	mg/100g	0.49 ± 0.03 d	2.83 ± 0.03 a
		%	0.01	0.01
	RS	mg/100g	0.18 ± 0.02 B	1.28 ± 0.06 A
		%	0.00	**0.01**
Total soluble sugars	SE	mg/g	41.85 ± 2.90 b	82.23 ± 3.12 a
		%	**67.23**	28.40
	RS	mg/g	39.95 ± 1.30 A	23.66 ± 1.17 B
		%	**65.35**	17.41
Soluble acid sugars	SE	mg/g	0.67 ± 0.27 b	1.88 ± 0.10 a
		%	1.08	0.65
	RS	mg/g	0.36 ± 0.01 A	0.24 ± 0.01 B
		%	0.59	0.18
% bioactive compounds refers to dry extract	SE	%	79.81	31.05
	RS		**97.69**	54.12
Cyanidin-3-glucoside	SE	µg/g	2.30	1.40
	RS	µg/g	1.20	11.73
Pelargonidin-3-glucoside	SE	µg/g	3.60	3.10
	RS	µg/g	N.D.	N.D.
Pelargonidin-3-rutinoside	SE	µg/g	4.80	2.10
	RS	µg/g	N.D.	N.D.
Quercetin-glucoside	SE	µg/g	38.10	5.70
	RS	µg/g	8.91	3.16
Ellagic acid derivative	SE	µg/g	42.30	1.30
	RS	µg/g	8.28	3.18
Ellagic acid	SE	µg/g	67.20	54.40
	RS	µg/g	9.39	7.27
Kaempferol-3-glucoside	SE	µg/g	38.40	2.10
	RS	µg/g	N.D.	N.D.
Kaempferol	SE	µg/g	7.2	0.0
	RS	µg/g	N.D.	N.D.
Antioxidantactivity				
DPPH	SE	mmol TE/kg fresh weight	5.33 ± 0.19 a	2.39 ± 0.15 c
	RS		2.67 ± 0.02 A	2.07 ± 0.02 B
FRAP	SE	mmol TE/kg fresh weight	335.60 ± 11.99	N.D.
	RS		263.68 ± 7.90 a	115.86 ± 6.54 b
ORAC	SE	mmol TE/kg fresh weight	37.89 ± 1.14 b	20.30 ± 1.26 c
	RS		19.70 ± 0.74 A	8.20 ± 0.29 B

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599 Table 6. Pearson correlation coefficients among the different variables studied for the
600 extracts obtained using the DES-7 solvent and acetone. The values marked in bold are
601 equal to or greater than 0.40 in absolute value.

	Total soluble phenolics	Soluble anthocyanins	Totalsoluble sugars	Soluble acid sugars	DPPH	FRAP	ORAC
STRAWBERRY EXTRUDATE							
Total soluble phenolics	**1.00**	**0.47**	**0.58**	**0.66**	**0.92**	**0.52**	**0.66**
Soluble anthocyanins	**0.47**	**1.00**	**0.62**	**0.96**	0.09	**-0.48**	0.37
Soluble sugars	**0.58**	**0.62**	**1.00**	**0.56**	0.38	0.16	-0.13
Soluble acid sugars	**0.66**	**0.96**	**0.56**	**1.00**	0.32	-0.29	**0.60**
DPPH	**0.92**	0.09	0.38	0.32	**1.00**	**0.80**	**0.58**
FRAP	**0.52**	**-0.48**	0.16	-0.29	**0.80**	**1.00**	0.10
ORAC	**0.66**	0.37	-0.13	**0.60**	**0.58**	0.10	**1.00**
RASPBERRY EXTRUDATE							
Total soluble phenolics	**1.00**	**0.95**	**-0.67**	**0.41**	**0.64**	0.31	0.23
Soluble anthocyanins	**0.95**	**1.00**	**-0.88**	0.10	0.35	-0.02	-0.09
Soluble sugars	**-0.67**	**-0.88**	**1.00**	**0.40**	0.14	**0.50**	**0.56**
Soluble acid sugars	**0.41**	0.10	**0.40**	**1.00**	**0.97**	**0.99**	**0.98**
DPPH	**0.64**	0.35	0.14	**0.97**	**1.00**	**0.93**	**0.90**
FRAP	0.31	-0.02	**0.50**	**0.99**	**0.93**	**1.00**	**1.00**
ORAC	0.23	-0.09	**0.56**	**0.98**	**0.90**	**1.00**	**1.00**

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